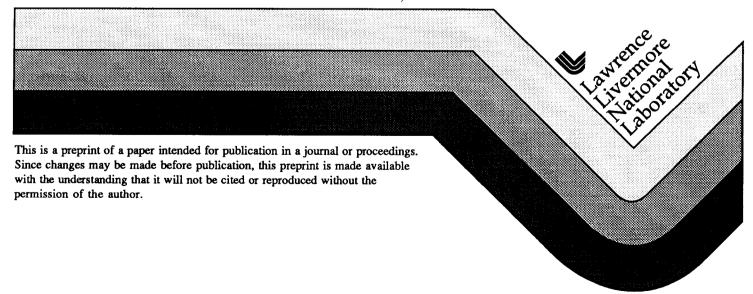
## Non-equilibrium Energy Loss for Very Highly Charged Ions in Insulators

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### Non-equilibrium Energy Loss for Very Highly Charged Ions in Insulators

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The energy loss of 144 keV Ar<sup>16+</sup> ions on a bilayer structure of C-CaF<sub>2</sub> has been measured. An asymmetry in the results is found depending on which layer is passed by the ion first: the energy loss is about four times larger when the CaF<sub>2</sub> layer is traversed by the ion first. We interpret this as an indication of the existence of a nonequilibrium charge state of the Ar ions inside the solid in the case of the insulator.

At present, it is unknown how long very slow (keV/u) highly charged ions (i.e. Ar16+, Kr34+, Th80+) can retain a non-equilibrium charge state in solids. In fact, there is no evidence that even the inner shell vacancies of such ions exist beyond the first few monolayers below the solid surface. Recently, it has been predicted that an increased nuclear-nuclear energy transfer, due to long range Coulomb interactions in insulators, is expected for these ions[1]. The object of our experiment is to demonstrate that such an effect actually occurs. This is accomplished through differential transmission energy loss spectroscopy using ion time of flight, as shown in Figure 1. Previous attempts to measure such energy loss effects in SiO2, through highly sensitive RBS measurements of the projected range of Xeq+, where 1≤q≤44, have produced negative results[2]. It is believed that this is due to the covalent nature of the insulator and therefore its screening of the ionic charge through strong polarization of the target atoms. A better candidate for the effect is a strong ionic solid such as MgO or CaF2. Furthermore, enhanced sensitivity to the effect would be achieved for transmission energy loss measurements through thin films, where the non-equilibrium state may exist over a large fraction of the ion's path length in the solid.

The differential technique is based upon the idea that the increased energy loss is expected only in ionic insulators (i.e. Cal?<sub>2</sub>) and not in metallic solids (i.e. C), since the valence electrons should efficiently screen the ion's potential at distances greater than the order of the lattice spacing. As depicted in Figure 2, a sandwich target structure is created under UHV conditions using thermal evaporation of several nanometers of CaF2 on a thin (10 nm) carbon foil backing, supported by a Cu TEM grid. Both AFM and TEM microscopic investigations indicate that the film is continuous for thickness of at least 2 to 4 nm. The transmitted energy loss spectrum of highly charged ions (i.e. Ar<sup>16+</sup>) incident on the ionic solid (i.e. CaF2) side of the two layer target structure is compared to that for ions incident on the metallic (carbon) side. Measurements of the charge state distribution of the ions transmitted through 100 nm C foils indicate that charge state equilibrium is achieved within the foil

(predominantly neutral and singly charged ions). Therefore, ions first passing through the C foil will interact with the CsF<sub>3</sub> film in equilibrium, whereas ions first entering the insulator will experience non-equilibrium effects. An asymmetry in the results for the two cases, therefore, indicates the existence of a non-equilibrium charge state inside the solid. It should be noted that, energy loss measurements of Ar16+ through various thickness of C fuils are in excellent agreement with equilibrium data[3], confirming one of the basic assumptions of the technique. However, a significant increase in the transited energy loss for highly charged ions incident on ionic insulating targets has been observed. In the case of 144 keVAr<sup>16+</sup> ions cm a 5 nm CaF<sub>2</sub> film, shown in Figure 3, the average energy loss for non-equilibrium charge states,  $\delta E_2$ , is a factor of four greater than that for the equilibrium charge state,  $\delta E_1$ , as achieved through initial transmission through a 10 nm C foil. Additionally, a significant increase in the energy loss straggling is observed for the case of highly charged ions in ionic solids. Both the increased energy loss and the enhanced straggling are thought due to the influence of reduced screening for small impact parameter collisions on energy transfer cross sections, due to hollow atom formation in the solid. These results represent evidence for the existence of very slow highly charged ions inside solids and suggest several techniques to study the formation and decay of hollow atoms in solids. Measurements of this asymmetry in the energy loss can be used to determine the average decay process of non-equilibrium charge states in solids.

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### Figure captions

Figure 1: Experimental arrangement for the time-of-flight measurements of transmitted energy loss for incident highly charged ions. The annular micro-channel plate (MCP) detector, located in front of the sample, is used to detect electrons emitted at ion impact and provide a start signal to the time-to-amplitude converter (TAC). The large area MCP detector located beyond the target provides the stop signal for the time-c)f-flight of the ion and has a position sensitive anode, allowing for measurements of the scattered angle-energy correlation. The flight path is 0.75 m with a solid angle for the PS-MCP detector of+/- 1.5 degrees.

Figure 2: A schematic representation of the experimental technique for the differential energy loss determination. First, the transmitted energy (time-of-fLight) is measured for the bare 10 nm C foil. Then a film of CaF<sub>2</sub> is deposited under UHV conditions. The transmitted energy is then measured for the ions incident on each of the two sides of the resulting hi-layer structure. The energy loss is determined directly from each side by subtraction of the bare C foil result,

Figure 3: Transmission time-of-flight results (converted into energy loss) for  $Ar^{16+}$  incident on the bare 10 nm C-foil ( $\Delta E_0$ , equilibrium) and the hi-layer structure: C-side first ( $\delta E_1 = \Delta E_1 - \Delta E_0$ ) and  $CaF_2$ -side first ( $\delta E_2 = \Delta E_2 - \Delta E_0$ ). A clear asymmetry exists between the results for the different incident sides of the same hi-layer structure, indicating non-equilibrium effects in the insulator.

